



Anthropogenic-driven perturbations on nitrogen cycles and interactions with climate changes

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Abstract

Anthropogenic activities have substantially perturbed the global nitrogen (N) cycle directly through enhancing reactive N (Nr) inputs and indirectly through climate and land-use change. However, the climatic impacts of the N cycle and its feedbacks on climate change remain very uncertain. In this review, we provide an overview of the dominant pathways by which anthropogenic Nr affects the climate system and summarize the available scientific assessments. We also review the latest progress on the responses of N cycle to changing climate to understand the potential for feedbacks between the N cycle and climate. With the urgent need to reduce Nr in the future to alleviate its negative environmental impacts, e.g. air pollution and eutrophication, we highlight the importance for bridging disciplines of atmospheric chemistry, ecology, and climatology to improve the scientific understanding and develop cobenefits for both environmental protection and climate change mitigation.

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Keywords

Anthropogenic nitrogen, Biogeochemistry–climate interactions, Climate effects of anthropogenic nitrogen, Terrestrial carbon–nitrogen interactions, Atmospheric nitrogen chemistry, Ammonium and nitrate aerosols, Nitrogen cycles under climate changes.

Introduction

Nitrogen (N) constitutes nearly 78% of the atmosphere; however, nearly 99% of atmospheric N is gas-phase molecular N (N_2) [1]. Two processes in the natural environment, i.e. biological nitrogen fixation (BNF) and lightning, can break the triple bond of N_2 and thereby

create reactive nitrogen (Nr). As one of the most important nutrients for living organisms supporting biosynthesis, growth, maintenance, and propagation, Nr can be subjected to a vast number of chemical and biochemical reactions and functions through its importance in enzyme synthesis and biomolecules [2,3]. However, Nr entering marine and terrestrial ecosystems may also be exported out of the biosphere and be emitted to the atmosphere, where it interferes with the radiative transfer in the atmosphere through tropospheric chemical reactions of greenhouse gases and aerosols [4,5]. As a result, anthropogenic Nr plays important roles in the global biogeochemical cycles (of carbon, nitrogen, phosphorus, etc.), further influencing environmental health [6,7], ecosystem biodiversity [8], and climate changes [9].

Fossil fuel combustion and N fixation technology (e.g. the Haber–Bosch reaction) have introduced large quantities of anthropogenic Nr to the Earth system since the beginning of the industrial revolution (around 1750). The scale of this perturbation has grown exponentially since the beginning of the industrial revolution and, in current decades, is estimated to be on par with or even exceed natural Nr generation through BNF and lightning [10]. Excessive Nr, on the one hand, substantially affects climate. For instance, increased nitrous oxide (N_2O) concentrations and ammonium (NH_4^+)/nitrate (NO_3^-) aerosol loadings in the atmosphere could warm and cool the atmosphere through the greenhouse effect and solar-radiation diffusion, respectively [11]. The enhanced Nr inputs into ecosystems through N deposition and fertilizer and manure application may alleviate global warming by increasing ecosystem carbon sequestration [12,13]. Furthermore, short-lived nitrogen oxides (NO_x) play vital roles in atmospheric chemistry and non-linearly alter atmospheric greenhouse gases of ozone (O_3) [14] and methane (CH_4) [15]. On the other hand, climate change induced by anthropogenic greenhouse gases also substantially affects almost all of the natural N processes [16,17], with the possibility of causing feedbacks to climate. The net climate effects of anthropogenic Nr as well as the N climate feedbacks, although understood in principle, are challenging to quantify and therefore remain purely constrained.

The importance of interactions between N dynamics and climate has gained recognition in the past decades [9,16,18]. Substantial efforts have been dedicated in the past years to improve the scientific understanding through field experiments, meta-data analysis, machine-learning techniques, and model simulations. In this

review study, we aim to give an overview on previous studies, most of which have predominantly focused on specific N compounds or processes within the global N cycles, and integrate the latest key findings with respect to interactions of N cycles and climate change. As illustrated in Figure 1, we will firstly overview the pathways that N cycles influence climate and identify the contributions led by direct anthropogenic Nr inputs (Section [Main climate-affecting N compounds](#)), then we will briefly demonstrate the recent discoveries of key N-cycle processes responses to changing climate as well as the anthropogenic perturbations (Section [Climate feedbacks due to perturbed N cycle](#)). Finally, we will conclude and outline future perspectives in Section [Concluding remarks](#).

Main climate-affecting N compounds

N₂O

N₂O is a long-lived greenhouse gas with a global warming potential that is about 273 times greater than that of CO₂ per unit mass over a period of 100 years. IPCC AR6 reported with high confidence that atmospheric N₂O mole fractions have risen from 270 ppb in 1750 to 331 ppb in 2019 and have contributed to $+0.21 \pm 0.03 \text{ W m}^{-2}$ effective radiative forcing (ERF) relative to the preindustrial period [19]. Increases in agricultural fertilizer/manure application, N deposition, and fossil fuel combustions were identified as primary drivers behind this increase of N₂O [20], but the exact attribution of anthropogenic contributions to the N₂O increases is still challenging. Anthropogenic sources during 2007–2016 were estimated to be 7.3 [4.2–11.4] Tg N yr⁻¹ among the global total 17.0 Tg N yr⁻¹

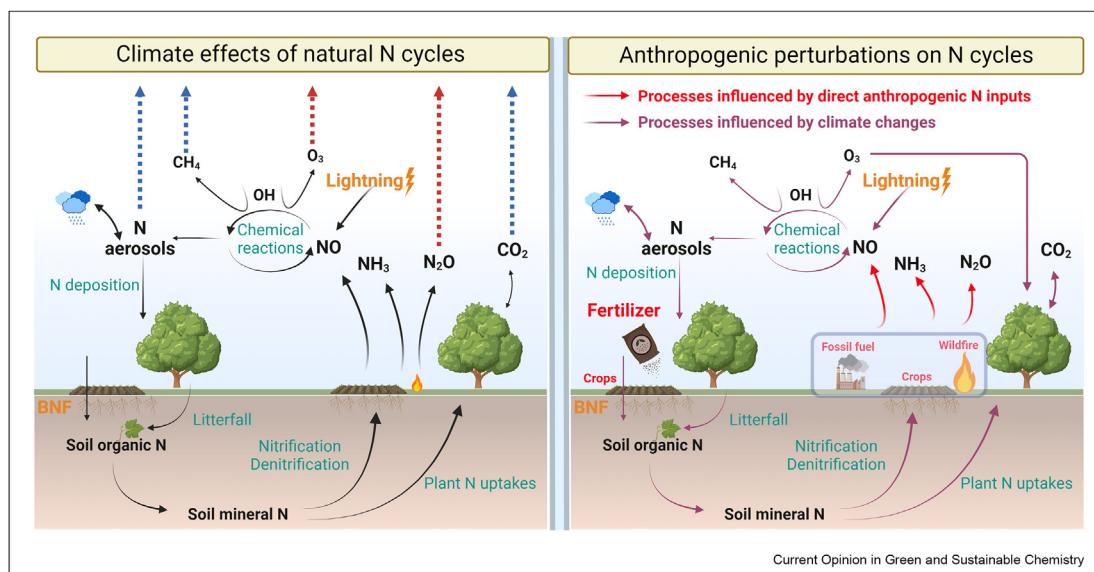
emissions [21]. Limited but also challenging flux measurements [22–24], uncertain and dynamic spatiotemporal distributions of N fertilizer application, and agricultural land uses [25], as well as imperfections in model representations [26] together are the main cause for this large range.

N-species aerosols

Aerosols remain in the troposphere for a timescale of days to weeks. Most compounds result in a cooling effect on climate as they scatter solar radiation and interact with cloud formation and lifetime. Ammonium (NH₄⁺) and nitrate (NO₃⁻) are the two dominant N-species aerosols. It is well-known that the major source of ammonia (NH₃), which is the primary precursor to generate NH₄⁺, is from agricultural activities [27], but significant gaps remain in quantifying global NH₃ emissions. Recent bottom-up estimates assessed global agricultural NH₃ emissions at about 58 Tg N yr⁻¹ in 2010 [28], whereas the top-down inversion accounting for natural and other anthropogenic NH₃ sources suggested global emissions from 90 to 191 Tg N yr⁻¹ [29,30]. Some studies also showed current emission inventories tended to underestimate NH₃ emissions particularly in hotspot regions, such as Western Europe [31] and Eastern China [32]. With uncertainties in NH₃ emissions, the ERF of NH₄⁺ in 2014 was estimated to be about -0.07 W m^{-2} , based on ensemble means of earth system models [11].

Emissions of nitrogen oxide (NO_x), which is the main precursor of NO₃⁻, are dominated by fossil fuel sources [27]. Both bottom-up and top-down estimates showed consistently similar magnitudes and decadal trends in

Figure 1



The climate effects of Nr compounds (left) also with anthropogenic-driven perturbations on N cycles (right). For each panel, the left part indicates the Nr inputs to the terrestrial ecosystem, whereas the right part indicates the Nr fluxes to the atmosphere. The blue or red dashed arrows on the top of the left panel indicate the cooling or warming effects of each component on climate, respectively. Abbreviation: Nr = reactive nitrogen.

anthropogenic NO_x emissions [33–35]. NO_x engages in numerous chemical reactions and significantly perturbs atmospheric hydroxyl radical (OH) concentrations (see below Section [NO_x indirect effects through atmospheric chemical reactions](#)). The non-linearity of atmospheric chemistry introduces uncertainties in the global burden, spatial distribution, as well as the particle sizes of NO₃⁻, all of which are essential to quantify the NO₃⁻ climate effects. Therefore, estimates of the cooling effects of NO₃⁻ through solar diffusion varied from very small ($\sim -0.025 \text{ W m}^{-2}$) to more substantial effects (-0.14 W m^{-2}) [11,36–39]. The interactions between NO₃⁻ aerosols and cloud remained quite uncertain, with radiative forcing assessed by a limited number of studies ranging from -0.05 to -0.22 W m^{-2} [40–42].

NO_x indirect effects through atmospheric chemical reactions

As the highly reactive gas, NO_x is involved in various chemical reactions with other green-house gases, in particular, O₃ and CH₄. It thereby affects climate indirectly by changing the lifetime and thus the atmospheric burden of these gases. In general, increased atmospheric NO_x concentrations will lead to higher OH concentrations, further increasing O₃ concentrations [14] while, at the same time, shortening CH₄ lifetime and thereby reducing CH₄ concentrations [15]. A recent assessment based on an ensemble of Earth System models showed that NO_x-induced O₃ enhancement warmed the climate by $+0.2 \pm 0.07 \text{ W m}^{-2}$, whereas the reduced CH₄ lifetime led by NO_x contributed a cooling effect of about -0.2 W m^{-2} to -0.37 W m^{-2} until 2014 since pre-industrial times [11]. The complexity of OH chemical dynamics and non-linear interactions among NO_x, volatile organic compounds, and O₃ is the primary cause for the uncertainty in this quantification.

C–N interactions in terrestrial ecosystems

The majority of the N element in the terrestrial ecosystem constitutes plants and microbes in organic forms and plays crucial roles in enzyme-mediated processes. However, the available N for plants is normally in the forms of dissolved inorganic N, such as ammonium or nitrate ions, which is mineralized from soil organic N by microbial activities [43]. Meanwhile, soil microbes also regulate BNF levels, thereby influencing the overall N availability for plants [44]. A portion of the mineralized inorganic N will be lost from the terrestrial ecosystem as gases phase through nitrification and denitrification [26], or as dissolved forms through leaching [45].

Next to phosphorus, the N in terrestrial ecosystems is one of the most important nutrients for plants and microbial organisms. It is essential for C assimilation, growth, and maintenance and thereby tightly connected to the C cycle. The N limitation of biomass production is generally believed to be strong in natural temperate

and boreal forests [46], attenuating the expected increases in carbon storage due to CO₂ fertilization of plant photosynthesis under future climate change [47]. Anthropogenic-driven increases in N fertilizer application and atmospheric Nr deposition can increase productivity by lifting N limitation. Synthesis of available data from ecosystem manipulation experiments by meta-analysis or machine learning has revealed responses of terrestrial C sinks to N addition, ranging from $1 \text{ kg C (kg N)}^{-1}$ to $25 \text{ kg C (kg N)}^{-1}$ depending on different land cover types and amounts and types of N addition [13,48–50].

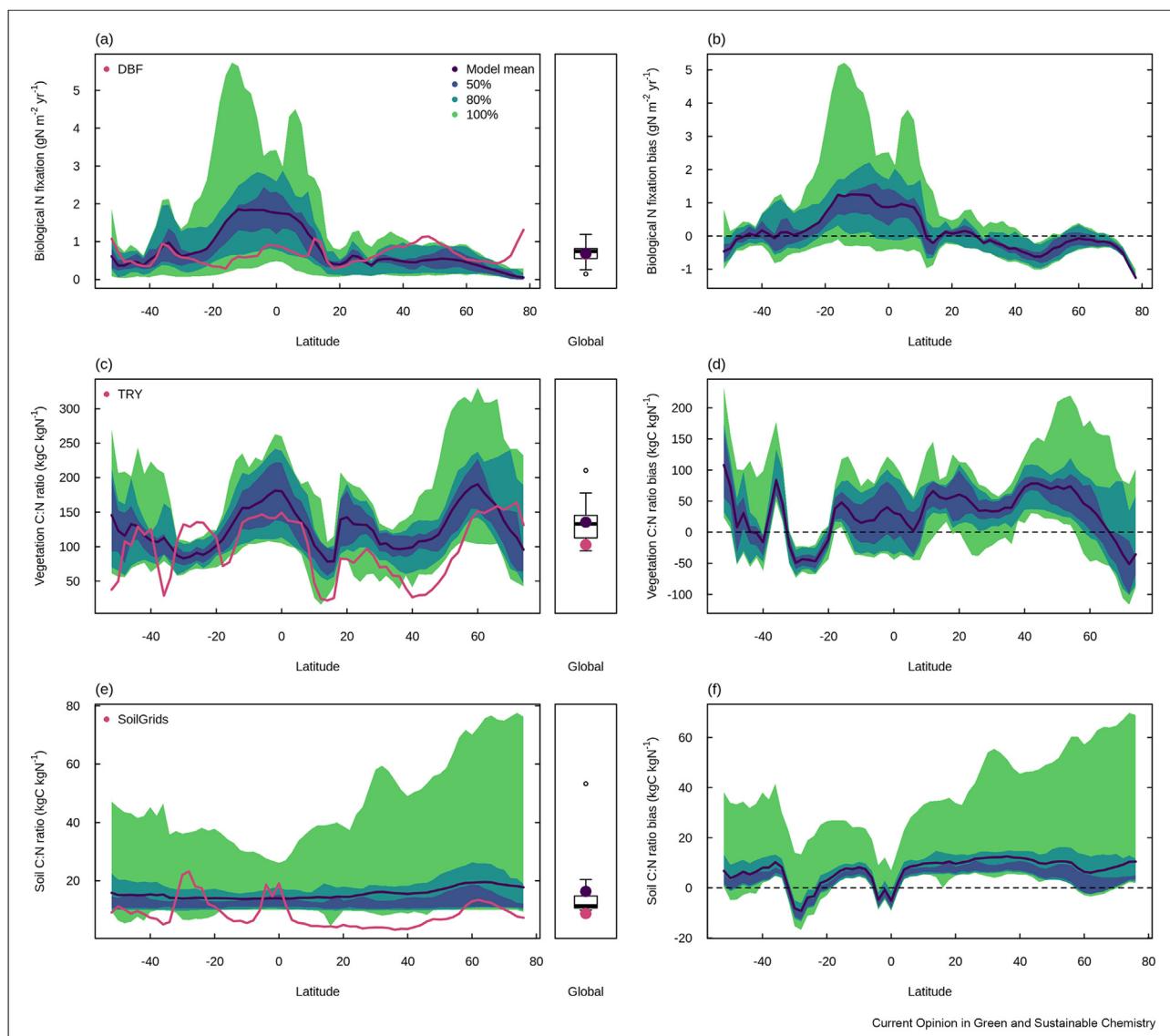
Terrestrial biosphere models (TBMs) integrate process understanding of biogeochemical cycles at large scales and are another useful tool to quantify the climate effects of N-induced increases in C sequestration [3]. For instance, Zaehle et al. [51] attributed a cumulative cooling effect of about -0.1 W m^{-2} to Nr-induced carbon storage in the terrestrial biosphere since pre-industrial times by applying a TBM to reproduce global observed trends of atmospheric CO₂. In recent years, dynamic C–N coupling has been induced in an increasing number of TBMs [52–55]. These models generally agree with earlier findings, but evaluation against independent benchmark highlights remaining uncertainty and a formidable challenge to further constrain model uncertainties to better quantify the N effects on terrestrial carbon sinks and the resulting climate impacts [53] (Figure 2).

Other Nr-related climate effects

There are additional, less well-quantified pathways of Nr known to affect climate, which are summarized here: (1) Nr-induced increases in aerosols and O₃ could influence terrestrial carbon sinks by reducing ratios of direct to diffuse photosynthetically active radiation [57,58] and by damaging leaf photosynthesis [59–61], respectively. (2) Nr inputs into terrestrial ecosystems may reduce soil CH₄ uptake [48,62] and thus increase atmospheric CH₄. However, soil CH₄ uptake plays only a small role in the global CH₄ cycle, and the magnitude of the Nr effects ranges from -60% to $+10\%$ [63]. (3) Riverine and marine N₂O emissions and the C–N interactions might also be influential on climate. Yao et al. [64] suggested that increased N inputs enhanced riverine emissions of N₂O from $70.4 \pm 15.4 \text{ Gg N yr}^{-1}$ in 1900 to $291.3 \pm 58.6 \text{ Gg N yr}^{-1}$ in 2016, whereas marine N deposition is estimated to contribute to present-day marine N₂O emissions by about $0.01\text{--}0.32 \text{ Tg N yr}^{-1}$ [19], and the marine primary productivity by 0.3 Pg C yr^{-1} increases [2] but with neglectable changes in the resultant air–sea CO₂ fluxes [65].

The net climate effects of anthropogenic Nr

Quantifying the net climate effects of anthropogenic Nr requires crossing different time scales and disciplines

Figure 2

Latitudinal distributions and global means of BNF, vegetation C: N ratio, and soil C: N ratio simulated by the TRENDY-N ensemble (averaged across models over 1980–2021) in comparison to observations. Panels (a, c, e) show the latitudinal distribution of the mean, and boxplots show the global mean. Panels (b, d, f) show the latitudinal distribution of the bias. Latitudinal distributions show the mean (black line) and the 50 %, 80 %, and 100 % percentiles across models. Boxplots show the median, interquartile range (box), and 80 % percentiles (whiskers) across models. Observation-based datasets are from Davies-Barnard and Friedlingstein [56] for biological N fixation, the TRY plant trait database for vegetation C:N ratio, and SoilGrids for soil C:N ratio. This figure is from Kou-Giesbrecht et al. [53]. Abbreviations: BNF = biological nitrogen fixation; TRY (TRY—not an acronym; Kattge et al., 2011 [107]).

among atmospheric chemistry, climate dynamics, and ecological science. To date, this gap among different scientific communities and quantifications is typically bridge in a ‘puzzle-style’ framework, wherein climate effects (e.g. radiative forcing) or key parameters (e.g. terrestrial C responses to N addition; N₂O emission factors) from previous independent studies are combined to generate a ‘best estimate’ of anthropogenic Nr climate effects. Table 1 summarized the four most recent and comprehensive estimates to our knowledge,

covering the U.S., Europe, China, and globe. To improve our understanding of the Nr imprint of anthropogenic Nr on climate, it is important to (1) address remaining uncertainties in the underlying regional to global emission databases for all Nr species and improve consistency between emission categories; (2) integrate process-understanding to derive non-linear response functions of ecosystems to reduce the dependency of the assessments on linear approximations; and (3) appropriately deal with the evaluation of climate

Table 1

Summary of present-day climate effects of anthropogenic Nr. The uncertainty ranges of each estimate were indicated by standard deviation (\pm) or square brackets depending on each individual study.

	Erisman et al. [9]	Pinder et al. [66]	Butterbach-Bahl et al. [67]	Shi et al. [68]
Region	Global	U.S.	Europe	China
Metrics	Radiative forcing	GWP20/GWP100	Radiative forcing ^a	GWP20/GWP100
Units	W m ⁻²	Tg eCO ₂	W m ⁻²	Tg eCO ₂
N ₂ O	+0.16	+314 [+202, +428]/ +291 [+180, +395]	0.017	345 ± 19/311 ± 17
Nr-related aerosols	-0.38	-36.67 [-8.18, -103.5]/0	-0.0165	-156 ± 45/-0.12 ± 0.13
NOx effects on CH ₄ lifetime		-271 [-177.4, -385]/	-0.0046	-349.8 ± 155.6/-13.2 ± 7.6
NOx effects on O ₃	+0.13	-7.19 [0, -15.64]	0.0029	
N-induced terrestrial carbon sinks	-0.2	-206.8 [-134.65, -292.4]/ -155.02 [-98.96, -236.76]	-0.019 (excluding agricultural fertilization)	-49.9 ± 54.7/-86.2 ± 39.7
Other effects	+0.05	[+66 + 140]	0.0453	111 ± 100/111 ± 100
Net climate effects	-0.24 [-0.5 + 0.2]	-200.47 [-299.6, +17.6]/ +128.79 [+112.2, +317.1]	-0.0173	-100 ± 414/322 ± 163

Abbreviation: Nr = reactive nitrogen.

^a The values indicated the global radiative forcing induced by emissions in Europe.

impact by accounting for regional and global short-lived as well as long-lived climate forcers.

Climate feedbacks due to perturbed N cycle

Besides direct anthropogenic Nr inputs, anthropogenic-driven climate change substantially alters the global N cycle, which in turn results in feedback effects on climate. Here, we provide a brief overview of key findings in the past years covering changes in ecosystem N cycles and atmospheric Nr-related chemistry and extrapolate the potential climate feedbacks.

Responses of ecosystem N cycling to climate changes

A number of processes in the terrestrial N cycle are substantially influenced by climate changes through the responses of enzyme activities, microbial species, and strategy [69]. Firstly, the BNF, the major natural source of Nr, is responding to temperature with new emerging evidence to better quantify the optimal temperature for BNF and its thermal acclimation to growth temperature [70,71]. Secondly, increased temperature accelerates the decay of organic material, which enhances inorganic N cycling in the soil and thereby enhance terrestrial carbon storage [72,73]. The intensification of inorganic N cycles can not only lead to a positive N₂O-warming feedback [74] but also intensify the soil emissions of NH₃ [75] and NO_x [76]. These processes may be particularly relevant in permafrost regions and peatlands, which are even more sensitive to global warming

due to arctic amplification. Recent studies have demonstrated that the increases in mineralized N with permafrost thawing failed to enhance the N availability of plants due to increased N demands and higher N-gas loss [77–79]. Thirdly, the simultaneous changes in multiple environmental factors, e.g. elevated CO₂ concentrations, warming, and changes in soil moisture, may enhance the strength of N limitation on terrestrial C uptake [80,81]. Last but not the least, land use changes and increased ecosystem disturbance (e.g. wildfire) may also deeply alter the whole ecosystem N cycle [82–84]. For example, the expansion of agricultural land may lead to higher N fertilizer application [85], whereas strategically intercropping of N-fixing crops (e.g. grain legumes) and cereals may alleviate such issues by improving N-use efficiency [86]. Meanwhile, more frequent wildfire under the changing climate accelerates the turnover rates of terrestrial N into the atmosphere, generating varied Nr-related gases and particles and substantially feeding back to the climate [87,88]. Overall, although specific components of the Nr-related feedbacks have been studied, a comprehensive and quantitative overview of the likely effects on future climate–biogeochemical feedbacks, such as N availability for the carbon cycle, is still lacking [19].

Responses of atmospheric Nr-related chemistry to climate changes

Higher temperatures in general not only accelerate reaction rates in atmospheric chemistry but also exert significant non-linear influences on individual reactions.

Clear evidence supports the hypothesis that higher temperature could accelerate evaporation of ammonium nitrate and thereby reduce Nr-related aerosol loadings [89]. Current Earth System models in general reported positive sensitivities of lightning NO_x emissions to the warmer climate [90], while insignificant or even negative sensitivities were reported in other studies [91,92]. The NO_x effects on CH₄ and O₃ with warming is strongly dependent on changes in global OH concentrations and distributions and therefore is subject to significant uncertainties [93,94].

Concluding remarks

In this review, we firstly summarized the known pathways in which Nr impacts climate. Recent findings underscored the vital roles of the N cycles in affecting global climate; however, each pathway was still associated with uncertainty. In particular, two major challenges of (1) improving estimates in global soil emissions of Nr gases (N₂O, NH₃, and NO_x) and (2) reducing uncertainties in atmospheric OH chemistry and associated NO_x effects on aerosols, O₃, and CH₄ substantially impede a more comprehensive understanding of the climatic effects of anthropogenic Nr. To fill this knowledge gap, on the one hand, long-term continuous observations of soil Nr-gas fluxes, especially for short-lived NH₃ and NO_x, and improved understandings in microbial dynamics with soil ammonification, nitrification, and denitrification (e.g. Ref. [95]) are both essential. On the other hand, new chemical mechanisms revealed from smog chambers (e.g. Ref. [96]), field campaigns (e.g. Ref. [97]), and the constraints by satellite retrievals (e.g. Ref. [98,99]) could promote a better description in global OH chemistry. Last but not the least, comprehensive Earth system models, which represent process-based terrestrial and marine N biogeochemical cycles, atmospheric chemical reactions, radiative transfer, and climate dynamics, which would be crucial to comprehensively reveal climate effects of anthropogenic Nr as well as the potential climate feedbacks through N cycles.

We further overviewed the latest progresses on the effects of anthropogenic perturbations on global N cycles. While the direct anthropogenic Nr addition into soil (e.g. via fertilizer application and N deposition) or atmosphere (e.g. via fossil fuel combustion) has been widely studied, the effects of indirect pathways, such as N processes influenced by changing climate, elevated CO₂, land-use changes, intensified wildfire, and diverse agricultural management strategies, remained inadequately understood. In-depth manipulating experiments in crucial zones, including tropical rainforest [100], permafrost [101], and agricultural lands [102], with as many N-cycle variables are urgently required. On the other hand, some national or regional practices have indicated that improved human management on

both forests and croplands holds promise in mitigating ecosystem disturbances (e.g. wildfire) [103], and increasing N use efficiency [104]. Those practices could increase Nr sequestration in the terrestrial plants and soil, thereby alleviating the environmental pollution led by anthropogenic Nr. For instance, the extensively practiced crop rotation, in particular over South Asia, with more N-fixing crops is effective in reducing Nr loss and increasing crop yields with less fertilizer application [105,106]. Nevertheless, it is still unknown how climate will respond to the efforts of reducing anthropogenic Nr. It is urgently required to bridge knowledge gaps among communities of agricultural nutrition, global biogeochemical science, atmospheric chemistry, and climate dynamics to achieve win-win in both environmental protection and climate change mitigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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